Studies on the Natural Smouldering of Cigarettes

III. Determination of the Thermal Properties of Tobacco Shreds Packed in Columns*

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INTRODUCTION

The basic thermal properties of a cigarette such as effective thermal conductivity, specific heat and thermal diffusivity play an important role in the temperature distribution or heating rate of tobacco shreds inside a smouldering cigarette (1).

The thermal properties of tobacco leaves have been studied to some extent previously (2–7) but mainly as a function of moisture content in connection with the curing or freeze-drying processes of tobaccos. There is little available information, however, in the literature on those of the cigarette itself. Such data are required for solving the heat-transfer problems of a smouldering cigarette.

For this reason, determinations of effective thermal conductivity and thermal diffusivity of tobacco shreds packed into columns simulating cigarettes were made on various types of tobaccos over a wide range of packing fraction. Results have shown that the effective thermal conductivity of tobacco shreds could be uniformly expressed by an experimental formula, as a function of the total void fraction inside the column regardless of tobacco types and stalk positions.

MATERIALS AND METHODS

Materials

Properties of the sample tobaccos are listed in Table 1. Tobacco leaves were cut into shreds of 0.65 mm wide, and an aliquot was ground to fine powder. After conditioning at 20 °C and 60% relative humidity, the shreds were used for the measurement of effective thermal conductivity and density, and the powders were used for the measurement of specific heat.

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Measurement of Effective Thermal Conductivity

A number of procedures is used for measuring the thermal conductivity of materials. Among these, the transient hot-wire method is especially useful for materials of low thermal conductivity. By this method, thermal conductivity can be easily measured in a short elapsed time using simply constructed equipment (8). The transient method was therefore selected to measure the effective thermal conductivity of the shreds packed in the simulated cigarette columns. A schematic diagram of the apparatus is shown in Fig. 1.

A nichrome wire (diameter: 0.1 mm, resistance: 1.428 ohm/cm) was used as a line heater, and a chromel-alumel thermocouple (diameter 0.1 mm) was firmly attached to the wire by spot welding. The heater wire was positioned along the central axis of a cylindrical vessel (30 mm diameter, 60 mm high) made of acrylic plastic, with holes for the thermocouple. The thermocouple junction was centered in the column as shown in Fig. 1. A specific quantity of conditioned, previously weighed tobacco shred was packed into the column from both ends as

Figure 1. Apparatus for the measurement of effective thermal conductivity of tobacco shreds.

Table 1. Properties of tobacco samples.

Tobacco sample		Moisture content (%, dry base)	Real density (g/cm ³)	Apparent density (g/cm ³)	Porosity (%)
Bright	A (lower)	12.9	1.427	0.763	46.53
	B (middle)	13.4	1.392	0.824	40.80
	C (upper)	12.7	1.371	0.869	36.62
Burley	D (lower)	11.9	1.448	0.695	52.00
	E (middle)	10.9	1.440	0.736	48.89
	F (upper)	11.5	1.422	0.730	48.66
Matsukawa	G (lower)	11.5	1.453	0.633	56.43
	H (middle)	11.9	1.386	0.691	50.14
	I (upper)	11.6	1.427	0.749	47.51

uniformly as possible. After packing, both ends were tightly stopped with a polyvinyl chloride plug containing a hole for the heater wire. The shred packed into the column was conditioned until the temperature of the heater wire reached the conditioning temperature (20 °C). Then, a constant electric current was passed through the heater wire for a few minutes using a D.C. current supply (Metronic Co., Model 592). The temperature rise of the heater wire was simultaneously recorded versus elapsed time.

The temperature rise of the heater wire depends on the effective thermal conductivity of the surrounding shred and on the heat produced in the heater wire as shown by the following equation (8):

$$\Delta T = (A_0/4\pi K_e) \ln(t_2/t_1) \quad [1]$$

where

$$\left. \begin{matrix} t_1 \\ t_2 \end{matrix} \right\} = \text{elapsed time } (t_2 > t_1, \text{ s}),$$

ΔT = temperature rise between t_1 and t_2 (°C),

A_0 = heat produced in the heater wire (cal/(cm s)),

K_e = effective thermal conductivity of shred packed into the column (cal/(cm s °C)).

According to equation 1, a plot of temperature versus time on semilog paper gives a straight line with a slope of $A_0/4\pi K_e$, and K_e can be calculated from the slope as given by the following equation:

$$\begin{aligned} K_e &= A_0/(4\pi S) \\ &= 0.239 (I/1000)^2 R/(4\pi S) \quad [2] \end{aligned}$$

where

S = slope of the straight line (°C),

I = electric current passed through the heater wire (mA),

R = resistance of the heater wire (ohm/cm).

Effective thermal conductivity was determined as a function of packing fraction. All measurements were conducted

at room conditions of 20 °C and 60% relative humidity, utilizing three sets of columns packed with shred. Measurements were repeated at least three times for each column, and the average was reported as the experimental value.

Measurement of Specific Heat

Specific heat was measured by means of a differential scanning calorimeter (DSC/Rigaku Denki Co., Model TG-DSC 8085-D1). Acceptability of this equipment for determining the specific heat of tobacco has been demonstrated by *Chakrabarti et al.* (6).

The calorimeter was scanned from 45 °C to 105 °C at a constant heating rate of 5°/min with a sensitivity of ± 2 mcal/s. The upper limit of the scanning temperature was selected by considering the thermostability of tobacco and stability of the base line obtained. Other details of the measuring procedure were essentially the same as those described by *Chakrabarti et al.* (6) except for the use of α -alumina as the reference material for specific heat instead of sapphire.

Determinations of specific heat were made not only on conditioned tobacco but also on bone-dry tobacco, on residual char left after the pyrolysis of tobacco (heated in helium up to 500 °C at a rate of 10°/min) and ash obtained by conventional methods (9). All experiments were repeated at least four times using two sets of sample pans.

Determination of Thermal Diffusivity

Thermal diffusivity ($[\alpha] = \text{cm}^2/\text{s}$) of shred packed into the column was calculated according to the following relation:

$$\alpha = K_e/(\rho_p C_p) \quad [3]$$

where

ρ_p = packing density of the shred packed into the column (g/cm³),

C_p = specific heat of shred (cal/(g °C)).

Determination of Packing Fraction and Measurements of Real and Apparent Shred Density

Packing fraction (1 - ε) of the shred packed into the column was determined according to the following equation:

1 - ε = (W/V) * (1/ρa), [4]

where

- ε = void fraction in the column,
- W = weight of the shred packed into the column (g),
- V = volume of the column (cm³),
- ρa = apparent density of the shred (g/cm³).

The real density ([ρr] = g/cm³) and apparent density of the shred were measured with a Beckman air pycnometer and by an oil-impregnation method reported by Masuo et al. (10), respectively. Then the porosity ([P] = %) of shred was given by:

P = 100(1 - ρa/ρr). [5]

The values of ρr, ρa and P are shown in the third, fourth and fifth columns of Table 1.

EXPERIMENTAL RESULTS AND DISCUSSIONS

Effective Thermal Conductivity

Applicability of the hot-wire method for measurement of effective thermal conductivity of tobacco shreds, as well as the experimental conditions and accuracy of the method, was first examined. Fig. 2 shows typical plots of temperature rise (ΔT) of the heater wire versus the logarithm of elapsed time (ln t) obtained with electrical currents of 60, 80 and 100 mA for the shred of Bright

Figure 2. Plots of temperature rise versus logarithm of elapsed time.

Sample: Bright-B Packing fraction: 0.34

Table 2. Comparison of observed and specified thermal conductivity.

Material	Observed value (10 ⁻⁴ cal/(cm s °C))	Specified value* (10 ⁻⁴ cal/(cm s °C))
Polyurethane foam	1.07	1.14
Silicone rubber	6.01	5.77

* Calibrated at EKO Instruments Trading Co., Ltd.

tobacco (B) with a packing fraction of 0.34. Each plot gave a straight line, satisfying the relation expressed by equation 1. However, if elapsed time were prolonged (even though such a case was not shown in Fig. 2), the linearity of the plot would be interrupted because of the finiteness of sample size. Therefore, it was considered preferable to determine the effective thermal conductivity within a short elapsed time. In this work, the effective thermal conductivity value was calculated from the slope ranging in elapsed time from 15 to 60 seconds by reference to a theoretical study (8) of the elapsed time range that would satisfy equation 1.

Fig. 3 shows the relationship between the electric current supplied to the heater wire and the effective thermal conductivity value, and also indicates the range of scatter in measured values in terms of the standard deviation. As can be seen from Fig. 3, the measured value was independent of the electric current within the range examined (40–140 mA), and the reproducibility of each value was invariably good. On the basis of these results, subsequent measurements were carried out under an electric current of 80 mA.

In addition, the accuracy of the measurement was checked by using plates (20 × 200 × 200 mm) of polyurethane foam and silicone rubber of known thermal conductivity (calibrated at EKO Instruments Trading Co., Ltd.). Table 2 compares the specified thermal conductivity values of these materials with values obtained by sandwiching the heater wire between mated halves of the sample. The measured and specified values agree within an error of less than 6%.

Figure 3. Relation between the electric current supplied and the value of effective thermal conductivity observed.

Sample: Bright-B Packing fraction: 0.34

Figure 4. Dependence of the effective thermal conductivity of tobacco shreds on the packing fraction.

It is clear from Figures 3 and 4 and Table 2 that the hot-wire method is applicable to the measurement of effective thermal conductivity of tobacco shred packed into columns and that the accuracy of the measurement is sufficient for practical use. The effective thermal conductivities of the conditioned sample tobacco shreds measured at 20°C as a function of packing fraction ($1 - \epsilon$) are summarized in Fig. 4. Without exception, the effective thermal conductivity rose with an increase in packing fraction. At the same packing fraction, shreds of Bright and upper leaves had higher effective thermal conductivities than other shreds. In contrast, these shreds tended to have lower porosities, as shown in Table 1. Such differences in effective thermal conductivity might be primarily dependent on the porosity of the shred or on the total void spaces inside the column, as will be demonstrated below.

It is well known that overall heat transfer in packed beds

Figure 5. Plot of $K_e/\epsilon_t^{1/3}$ versus $(1 - \epsilon_t^{2/3})/\epsilon_t^{1/3}$.

or porous materials arises not only from conduction in a solid itself but also from conduction, radiation and convection in void spaces. Therefore, effective thermal conductivity comprehensively includes the combined mechanisms of heat transfer in both phases. Kunii (11) presented the following equation, which made it possible to interpret the influences of void fraction and temperature on the effective thermal conductivity of highly porous materials:

$$K_e = (1 - \epsilon^{2/3}) K_s C + \epsilon^{1/3} (1 + \frac{2}{3} h_r D_p / K_g) K_g, \quad [6]$$

where

- K_s } = thermal conductivity of solid and gas
- K_g } = (cal/(cm s °C)),
- h_r = heat transfer coefficient of radiation
- = (cal/(cm² s °C)),
- C = correction factor,
- D_p = average diameter of void space (cm).

Since heat transfer by radiation may be negligibly small at a low temperature in the vicinity of room temperature, equation 6 can be simplified as follows:

$$\frac{K_e}{\epsilon^{1/3}} = \frac{(1 - \epsilon^{2/3})}{\epsilon^{1/3}} K_s C + K_g. \quad [7]$$

Equation 7 suggests that a plot of $K_e/\epsilon^{1/3}$ versus $(1 - \epsilon^{2/3})/\epsilon^{1/3}$ gives a straight line with a slope of $K_s C$ and an intercept of K_g . Tobacco shred packed into a column can be considered a type of porous material. Therefore, by replacing the void fraction (ϵ) inside the column packed with shreds with the total void fraction (ϵ_t) which corresponds to the void fraction of porous material defined by equation 8 (i.e. by considering the pores of the shreds also to be void), the experimental results shown in Fig. 4 were rearranged according to equation 7. The results of these calculations are shown in Fig. 5.

$$\epsilon_t = 1 - (1 - \epsilon) \frac{Q_a}{Q_r} = 1 - \frac{W}{V} \cdot \frac{1}{Q_r}. \quad [8]$$

It can be seen from Fig. 5 that experimental data are plotted on a single straight line with a slope of $K_s C =$

Figure 6. Relation between specific heat and temperature.

$7.56 \cdot 10^{-4}$ and an intercept of $K_g = 1.08 \cdot 10^{-4}$. Thus, the effective thermal conductivity of conditioned shreds packed into the column was uniformly expressed as a function of total void (ϵ_t) regardless of tobacco types and stalk positions, as given by equation 9:

$$K_e = 7.56 \cdot 10^{-4} (1 - \epsilon_t^{2/3}) + 1.08 \cdot 10^{-4} \epsilon_t^{1/3} \quad [9]$$

The value of K_g obtained was approximately 1.7 times greater than that of thermal conductivity of air, suggesting that the heat transfer in the void would be greater than that expected from the thermal conductivity of air probably because of convection, mass transfer of vapour, etc. in the void space.

Although the effective thermal conductivity values obtained in this work could not be directly compared with data previously presented by Samfield et al. (4) because of the lack of void fraction data in their paper, both values were of the same order of magnitude.

Specific Heat

Dependence of specific heats of various kinds of tobacco, char, and ash on temperature are shown in Fig. 6. The specific heat of every sample examined increased almost linearly with temperature, especially for tobacco. Bright tobacco showed appreciably higher specific heat than air-cured types (Burley, Matsukawa), while specific heats of char and ash seemed to be no longer dependent on the original tobacco type. The specific heat of dried Bright tobacco agreed very closely with that reported by Chakrabarti et al. (6), and the dried Burley and Matsukawa tobaccos were in fair agreement with those of the dried woods (12) given by equation 10:

$$C_p = 0.266 + 0.0116 T \quad [10]$$

where T = temperature ($^{\circ}\text{C}$).

On the other hand, specific heat of each conditioned tobacco was considerably higher than that calculated by the "method of mixture" (6), similarly to the results reported by Brock et al. (3) and Chakrabarti et al. (6).

Thermal Diffusivity

Thermal diffusivity at 20°C of the conditioned shred packed into the column was calculated using the values of effective thermal conductivity, specific heat and packing density. The value of specific heat at 20°C used for the calculation was estimated by extrapolating the linear plot of the specific heat of conditioned tobacco versus temperature shown in Fig. 6.

Plots of thermal diffusivity versus packing fraction ($1 - \epsilon$) were prepared for each tobacco shred sample. Typical plots are shown in Fig. 7. It became apparent that the packing fraction exerted a pronounced influence on the thermal diffusivity, and at the same packing fraction, shreds of air-cured types had higher thermal diffusivities than flue-cured types.

Until this study was conducted, thermal properties of tobacco shreds packed into columns had been studied mainly as a function of packing fraction. Results obtained through this study permit the evaluation of the thermal

Figure 7. Dependence of the thermal diffusivity of tobacco shreds on the packing fraction.

Table 3. Thermal properties at 20 °C of conditioned shreds at a packing fraction of 0.34.

Tobacco		Packing density (g/cm ³)	Effective thermal conductivity (10 ⁻⁴ cal/(cm s °C))	Specific heat (cal/(g °C))	Thermal diffusivity (10 ⁻³ cm ² /s)
Bright	A	0.259	1.91	0.49	1.50
	B	0.280	2.07	0.47	1.57
	C	0.295	2.09	0.50	1.41
Burley	D	0.236	1.88	0.41	1.94
	E	0.250	1.96	0.42	1.87
	F	0.248	1.86	0.43	1.74
Matsukawa	G	0.215	1.79	0.43	1.94
	H	0.235	1.88	0.43	1.86
	I	0.255	2.02	0.44	1.80

properties of a cigarette, which are useful for solving certain heat-transfer problems occurring inside a smouldering cigarette.

The packing fraction of tobacco shred is not widely different among commercially available cigarettes, generally in the range of 0.30–0.36. Therefore, the values of effective thermal conductivity and thermal diffusivity at 20 °C of the conditioned shred samples at a packing fraction of 0.34 are summarized in Table 3, together with extrapolated specific heats and packing densities.

SUMMARY

The values of effective thermal conductivity, specific heat and thermal diffusivity of various kinds of tobacco shreds, which are necessary for the solution of heat transfer problems of a smouldering cigarette, were determined.

Effective thermal conductivity of tobacco shred packed into simulated cigarette columns was measured by a transient hot-wire method as a function of packing fraction at conditions of 20 °C and 60% relative humidity. Specific heat was measured with the aid of a differential scanning calorimeter (DSC). The thermal diffusivity was obtained from the values of effective thermal conductivity, specific heat and packing density.

Effective thermal conductivity increased with increasing packing fraction ($1 - \epsilon$) and was uniformly expressed as a function of the total void fraction (ϵ_t) inside the column regardless of tobacco types and stalk positions, whereas thermal diffusivity decreased with the packing fraction.

At the same packing fraction, Bright shreds produced higher values of effective thermal conductivity as well as higher specific heats (although specific heat was independent of the packing fraction) but had lower thermal diffusivity values than Burley and Matsukawa shreds.

ZUSAMMENFASSUNG

Es wird über die Bestimmung der effektiven Wärmeleitfähigkeit, der spezifischen Wärme und der Temperaturleitfähigkeit verschiedener Arten von Tabakfasern berichtet. Die Kenntnis dieser Größen ist notwendig zur

Lösung des Problems des Wärmetransports in einer glimmenden Zigarette.

Die effektive Wärmeleitfähigkeit wurde an Tabakfasern, die sich in Zigaretten simulierenden Zylindern befanden, in Abhängigkeit von der Stopfdichte mit Hilfe einer Hitzdrahttechnik bei der Temperatur von 20 °C und einer relativen Luftfeuchtigkeit von 60% gemessen. Die spezifische Wärme wurde mit Hilfe der quantitativen differentiellen Thermoanalyse (DSC) bestimmt. Die Temperaturleitfähigkeit wurde von den Werten der effektiven Wärmeleitfähigkeit, der spezifischen Wärme und der Stopfdichte abgeleitet.

Die effektive Wärmeleitfähigkeit stieg mit zunehmender Stopfdichte ($1 - \epsilon$) an und wurde für alle Tabakarten und Blattpositionen einheitlich in Abhängigkeit von dem gesamten anteiligen Leerraum (ϵ_t) im Zylinder ausgedrückt, wohingegen die Temperaturleitfähigkeit mit der Stopfdichte abnahm.

Bei gleicher Stopfdichte hatten Fasern von Bright-Tabak eine größere effektive Wärmeleitfähigkeit und auch eine höhere spezifische Wärme (obwohl diese von der Stopfdichte nicht abhängig war), andererseits jedoch eine niedrigere Temperaturleitfähigkeit als Fasern von Burley- und Matsukawa-Tabak.

RÉSUMÉ

On a déterminé la conductibilité thermique effective, la chaleur spécifique et le coefficient de diffusion thermique de différentes sortes de scaferlati, nécessaires au calcul du transfert de chaleur dans une cigarette en combustion libre.

La conductibilité thermique effective du scaferlati dans les boudins de cigarettes simulées a été mesurée par une méthode à fil chaud transitoire en fonction du bourrage, à 20 °C et 60% d'humidité relative. La chaleur spécifique a été mesurée au moyen d'un calorimètre différentiel à faisceau baladeur (DSC). Le coefficient de diffusion thermique a été obtenu à partir des valeurs de conductibilité thermique effective, de la chaleur spécifique et de la densité apparente.

La conductibilité thermique effective augmentait avec le bourrage ($1 - \epsilon$) et était exprimée systématiquement comme une fonction de l'espace vide fractionnaire total (ϵ_t) dans

le boudin indépendamment du type de tabac ou de la position des feuilles sur la tige, tandis que le coefficient de diffusion thermique diminuait avec le bourrage.

Au même bourrage, le scaferlati de tabac Bright produisait des valeurs de conductibilité thermique effective et une chaleur spécifique supérieures (bien que la chaleur spécifique ne dépende pas du bourrage), mais avait des coefficients de diffusion thermique inférieurs à ceux trouvés dans les scaferlatis Burley et Matsukawa.

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